Response to Comments of Reviewer #1

(comments in *italics*)

Manuscript number: acp-2021-119

Title: Impacts of aerosol-photolysis interaction and aerosol-radiation feedback on surface-layer ozone in North China during a multi-pollutant air pollution episode

Yang et al. examined the impacts of aerosols on surface ozone through the two well-known pathways, i.e., aerosol-photolysis interaction and aerosol-radiation feedback. The novelty of this study is its focus on the polluted episodes with elevated both $PM_{2.5}$ and ozone levels over North China. They also quantified the chemical and physical processes that drive the aerosol-radiation interactions.

Overall, this is a timely study and it clearly demonstrates the impacts of aerosols on ozone pollution. The structure of this manuscript is easy to follow. Although some of the manuscript needs further clarification, the results are generally convincing. As such, I think it is publishable after the following issues are addressed.

Response:

Thanks to the reviewer for the valuable comments and suggestions which are very helpful for us to improve our manuscript. We have revised the manuscript carefully, as described in our point-to-point responses to the comments.

Specific Comments:

1. *In Abstract: ozone changes refer to MDA8 ozone or daytime ozone?*

Response:

The ozone changes in abstract is daytime ozone. According to the reviewer's comments, we have added this information in the revised manuscript. (**Page 2, Line 33**)

2. *Line 177: a correlation coefficient of 0.66 reads like not high!*

Response:

According to the reviewer's suggestion, we have corrected it in the revised manuscript as follows: "The model can also reasonably capture the temporal variations of observed $PM_{2.5}$ and O_3 with correlation coefficients (R) of 0.66 for $PM_{2.5}$ and 0.86 for O_3." (**Page 7, Line 180-183**)

3. *Lines 179-181: the oxidation of SO_2 by NO_2 in aqueous aerosols is important for summertime?*

Response:

Thanks for your suggestion, we have changed the explanation in the revised manuscript as follows: "The failure to reproduce $PM_{2.5}$ peak values may be attributed to incomplete treatments of chemical reactions in WRF-Chem, e.g., missing the heterogeneous chemistry in the model (Cheng et al., 2016) and the lack of secondary organic aerosols (SOA) formation pathways in the aerosol module (Chen et al., 2019)." (**Page 7-8, Line 183-188**)

4. *Lines 248-251: this statement looks reasonable here, but in the later text the process analysis shows that chemistry will be enhanced by ARF. Instead, ARF decreases ozone through physical processes.*

Response:

Thanks for the reviewer's suggestion. We have deleted this sentence in the revised manuscript.

5. *Line 260: "is" should be "are". Please do proof-reading throughout the text.*

Response:

This sentence has been deleted in the revised manuscript. According to the reviewer's comments, proof-reading has been conducted through the whole revised manuscript.

6. *Line 310: It is Okay to use model levels (e.g., 12 levels), but it will be better to add model height in meters as well.*

Response:

Thanks for reviewer's suggestion. We have added the model height in meters in the revised manuscript. (**Page 12, Line 314, Line 317-319, Line 328**)

7. *Lines 326-327: why do you need this statement?*

Response:

Analyzing Fig. 8c we can conclude that ARF promotes the O_3 chemical production with a positive mean value of 0.66 ppb h^{-1}. The enhanced O_3 precursors due to ARF can promote the chemical production of O_3. According to the reviewer's comment, we have deleted this statement in the revised manuscript.

8. *Lines 327-328: Please provide evidence to support this conclusion.*

Response:

The typical VOCs/NO_x ratio is calculated to classify sensitivity regimes and to indicate the possible O_3 responses to changes in VOCs and/or NO_x concentrations. O_3 production is VOC-limited if the ratio is less than 4, and it is NO_x-limited if the ratio is larger than 15 (Edson et al., 2017; Li et al., 2017). The ratio of VOCs/NO_x ranging around 4-15 indicates a transitional regime, where ozone is nearly equally sensitive to each species (Sillman, 1999). As shown in Fig. R1(a-c), O_3 are mainly formed under VOC-limited and transition regimes in CAPAs, which means that the increased concentrations of VOCs and NO_x are favorable for ozone chemical production. As shown in Fig. R1(e) and (h), both the surface concentrations of VOCs and NO_x are increased when the impacts of ARF are considered. Thus, the contribution of CHEM in NOAPI is larger than that in NOALL. Similar results can also be found in Gao et al. (2018).

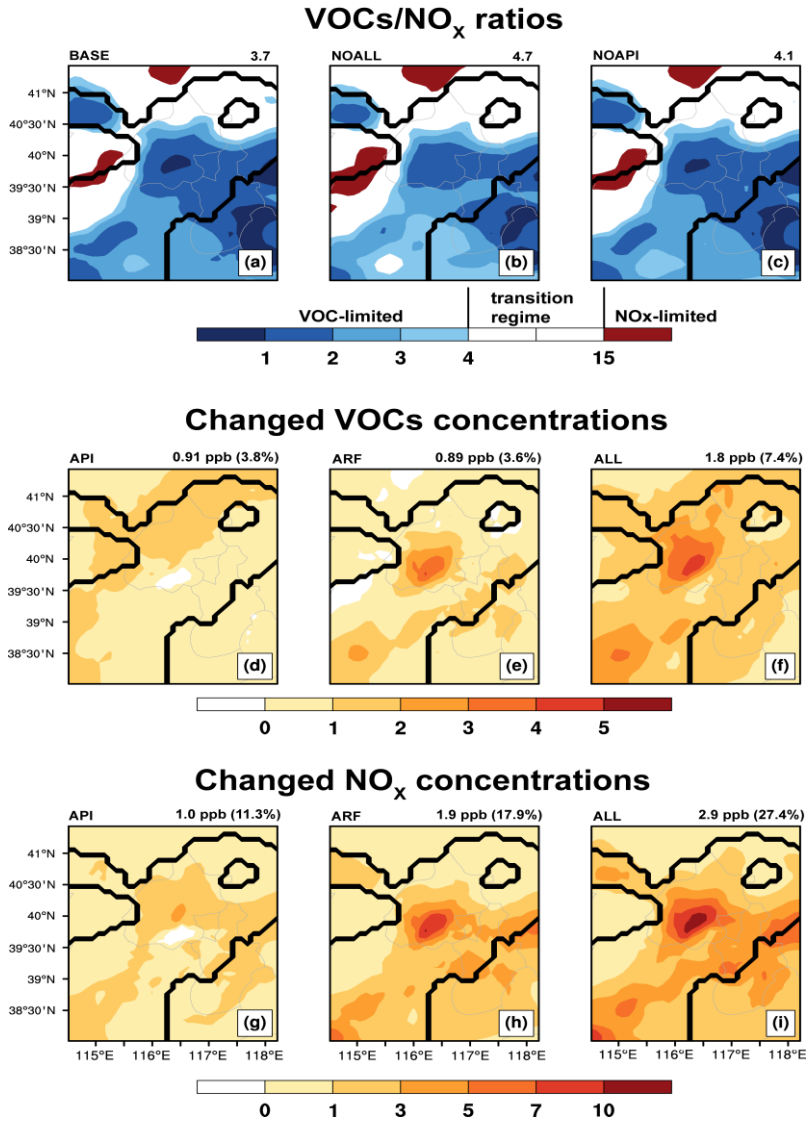

Figure R1. The ratios of VOCs/NO_x calculated from (a) BASE, (b) NOALL, and (c) NOAPI. The changed surface-layer concentrations of VOCs and NO_x (NO_2+NO, ppb) caused by (d, g) API, (e, h) ARF, and (f, i) ALL during the daytime (08:00-17:00 LST) from 28 July to 3 August 2014. The calculated values averaged over CAPAs are also shown at the top of each panel.

9. *Discussion. I think the authors should do some comparisons between your results with previous studies. This is important for readers to better understand your case study results. Moreover, how about the applicability of the calculated ROP of -0.14 ppb ($\mu g\ m^{-3}$)$^{-1}$?*

Response:

According to the comments of Reviewer#2, another two complex air pollution episodes (8-13 July 2015 and 5-11 June 2016) in this region are selected to conduct simulations for generating general conclusions (**Page 13-14, Line 343-365**). Meanwhile, a discussion about the impacts of secondary organic aerosols (SOA) is also added in the section 6 (**Page 15, Line 402-412**).

Thanks to the reviewer's comments. As the relationship between O_3 and $PM_{2.5}$ is non-linear, and the simple index of ROP can not fully represent the impacts of aerosols on surface O_3, so we

delete the ROP in the revised manuscript.

10. Fig.2: It will be better to add error bars for observed $PM_{2.5}$ and ozone.

Response:

According to the reviewer's suggestion, error bars have been added in Fig. 2 in the revised manuscript. (**Page 27**)

11. Fig.3: what are the cities these plots for?

Response:

The averaged T_2, RH_2, and WS_{10} are collected from ten meteorological observation stations, and the detail information about the sites is listed in Table S1. The photolysis rates of NO_2 ($J[NO_2]$) are observed in Peking University. More details are explained in section 2.3. (**Page 6**)

12. Fig.7: what are the layers your process analysis applied for? I don't see this key information here, as well as in the text.

Response:

The surface-layer, namely, first-layer O_3 concentrations are analyzed in Fig. 7. Thanks for reviewer's suggestion, we have added this information in the revised manuscript. (**Page 10, Line 266**)

Reference:

Chen, L., Zhu, J., Liao, H., Gao, Y., Qiu, Y., Zhang, M., Liu, Z., Li, N., and Wang, Y.: Assessing the formation and evolution mechanisms of severe haze pollution in the Beijing–Tianjin–Hebei region using process analysis, Atmos. Chem. Phys., 19, 10845–10864, https://doi.org/10.5194/acp-19-10845-2019, 2019.

Cheng, Y., Zheng, G., Chao, W., Mu, Q., Bo, Z., Wang, Z., Meng, G., Qiang, Z., He, K., and Carmichael, G.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, Science Advances, 2, https://doi.org/10.1126/sciadv.1601530, 2016.

Edson, C. T., Ivan, H.-P. and Alberto, M.: Use of combined observational- and model-derived photochemical indicators to assess the O_3-NO_x-VOC System sensitivity in urban areas, Atmosphere., 8, 22. https://doi.org/10.3390/ atmos8020022, 2017.

Li, K., Chen, L., Ying, F., White, S. J., Jang, C., Wu, X., Gao, X., Hong, S., Shen, J., Azzi, M. and Cen, K: Meteorological and chemical impacts on ozone formation: a case study in Hangzhou, China, Atmos. Res., 196, https://doi.org/10.1016/ j.atmosres.2017.06.003, 2017.

Sillman, S.: The relation between ozone, NO_x and hydrocarbons in urban and polluted rural environments, Atmos. Environ., 33, 1821-1845, https://doi.org/ 10.1016/S1352-2310(98)00345-8, 1999.

Thank you very much for your comments and suggestions.